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# Effects of High Intensity Electrical, Magnetic, Ultrasonic and Microwave Fields Upon the Microstructure, Processing and Properties of Metal and Ceramic Alloys



H.I. Aaronson
S.C. Sanday

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The effects of high-intensity electrical, magnetic, ultrasonic, and microwave fields upon the microstructural and mechanical properties of metal and ceramic alloys and the implications on the processing conditions for the forming components to near-net-shape are reviewed and discussed. Potentially significant developments that would take advantage of the most prominent effects in the forming of some components of interest to the DoD are identified.

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## CONTENTS

# EFFECTS OF HIGH-INTENSITY ELECTRICAL, MAGNETIC, ULTRASONIC AND MICROWAVE FIELDS UPON THE MICROSTRUCTURE, PROCESSING AND PROPERTIES OF METAL AND CERAMIC ALLOYS

## I. INTRODUCTION

This report is based in good part upon the unpublished proceedings of a symposium held at North Carolina State University, Raleigh, NC on July 17 & 18, 1989 under Army Research Office sponsorship on the effects of high intensity electric, magnetic, ultrasonic and microwave fields upon the response of metal and ceramic alloys to processing by deformation and heat treatment. Relevant material subsequently published, however, has also been included in this report.

Much of the early and also current work in this area was performed in the former USSR and has accordingly been viewed by many Westerners with considerable suspicion because of previous Soviet scientific "stumbles", e.g., the Rehbinder Effect and polywater. Indeed, the theoretical interpretations of many of the phenomena described in this report (many of which were provided by American physicists) are severely criticized by Prof. Doris Kuhlmann-Wilsdorf (Univ. of Virginia). However, certain of the technological achievements demonstrated, which have been confirmed and extended by American researchers, are both striking as such and also appear to have considerable potential military and industrial importance. Advantage can be taken of certain of these achievements, as will be briefly suggested in Section III of this report, to enable or ease the manufacturing of a wide variety of components for military and civilian applications in a cost effective manner, even though the theoretical bases for the electric and ultrasonic field and high density current effects remain incomplete. Salient among these are: wrought, rather than cast, gas turbine blades, carbon seal support rings, and other high temperature engine components from titanium aluminide alloys; missile nozzles, kinetic energy penetrators for gatling guns, shaped charge liners, and other components from tungsten alloys; and several other components from monolithic or powder form of Alloy 718, Ti-6Al-4V, and Fe-Co used in many DoD applications.

The following plan has been utilized in compiling this report. In the next section (II), the main achievements reported in modifying the microstructures and mechanical properties of materials by the application of fields are summarized successively by field type. The theoretical explanations which have been developed for the effects of each field are discussed immediately after its experimental consequences have been described. Suggestions and recommendations for efforts that could further the understanding of the observed phenomena have been added where appropriate. Section III is devoted to a brief overview of the potentially significant developments that

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would take advantage of the most prominent achievements described in Section II in the near-net-shape forming of components of interest to the DoD.

# II. ACHIEVEMENTS AND PROBLEMS OF HIGH-FIELD TREATMENTS

## A. Electric Current Passed Through Workpiece

### *1. Experimental Observations*

#### a. History

The first publication in this area appears to have come from the former USSR and much of the subsequent work was generated there. More recently, however, an increasing proportion of the research papers have emanated from the US as the possibilities, both scientific and industrial/military, have become apparent. The initial work, published in 1969, was that of Troitskii (1). He passed DC (direct current) pulses from $10^3$ to $10^7$ A/cm$^2$ with a duration of 100 μs through Zn single crystals undergoing plastic deformation. As shown in Fig. 1, a sharp load drop, of about 40%, resulted, but none occurred either during prior elastic deformation or subsequently following relaxation of the load or in the absence of current. The load drop increased with current density.

#### b. Plastic Deformation

Conrad, Sprecher, Cao and Lu (2) applied single DC 100 μs pulses of $10^5$ A/cm$^2$ to Zn single crystals and to various metal polycrystals in the course of experiments designed so as to avoid or minimize Joule heating, pinch and magnetostrictive "side effects". They found that the ratio of the strain rate, $\dot{\varepsilon}$, in the presence of electric pulses to that in the absence of pulses, $\dot{\varepsilon}_o$, is given by:

$$(1) \qquad \dot{\varepsilon}/\dot{\varepsilon}_o = (j/j_c)^n$$

where j is the applied current, $j_c$ is the critical current, (i.e., the minimum required to produce the "electroplastic effect", found to be $10^3$ - $10^4$ A/cm$^2$ for the Zn specimens), and n ~ 3. Figure 2 presents experimental data from which Eq. (1) was derived. Exploring the effects of the electron density associated with various elements upon the critical current, they showed that:

$$(2) \qquad j_c = A n_e^p$$

where A is a constant, $n_e$ is the electron density and p = 2/3 at 300 K and 2/5 at 77 K. Figure 3 displays some of the experimental results used to obtain Eq. (2).

Conrad et al. (2) reported that DC current expedited stress relaxation in Cu and Al. At 0.5 $T_m$, stress relaxation was much faster than at 0.3 $T_m$ for both metals, where $T_m$ is the absolute melting temperature. TEM (transmission electron microscopy)

showed that DC partially destroyed the dislocation cell structure usually associated with plastic deformation at temperatures high enough so that some diffusional relaxation of the dislocation structure through which the deformation process was accomplished can occur. Smaller effects were observed for AC (alternating current) than for DC of the same magnitude.

Even though the DC was "on" only $2 \times 10^{-4}$ of the processing time, a $10^4$ A/cm$^2$ current, applied in 100 μs pulses at a frequency of 2Hz, yielded a 2-3X increase in the fatigue life of polycrystalline Cu during rotating bending tests (2).

The creep rate of $V_3Si$ was increased by a DC of 2500 A/cm$^2$. Application of a similar AC current density exerted smaller effects upon creep kinetics (2).

Polarity effects were often observed. For example, when Boiko et al (4,5) compressed unicrystalline spheres of Cu, Al and W between parallel plates and $10^7$ A/cm$^2$ DC was applied for 0.01 sec., the contact area produced at the positive pole was larger on the Al and Cu spheres and the reverse obtained for the W spheres. This difference was ascribed to the circumstance that the first two metals are n-type conductors whereas W is a p-type conductor. Zuev et al. (6) measured the velocity of {11-22}<11-23> dislocations in Zn crystals as a function of externally applied stress with the etch pit technique. When single pulses of $7.5 \times 10^3$ A/cm$^2$ were applied for 200 μs, the dislocation velocity was increased in both the parallel and the antiparallel directions, but the increase in the parallel direction was larger, indicating a vectorial effect of the current.

The effects described in this subsection have been utilized to improve industrially oriented mechanical metallurgical processes. One of these is wire drawing. Troitskii et al. (7) mainly tested Cu, W and stainless steel wires. Current densities in the range from $10^4$ to $3 \times 10^5$ A/cm$^2$, pulse durations from 3 to 100 μs and pulse frequencies of 100 to 30,000 Hz were utilized. Drawing speeds varied from 5 to 300 m/min; reductions in area of 4 to 20% were employed; original wire diameters were usually 50 -100 μm (though in one set of experiments wire diameters varied from 150 to 300 μm). Both continuous and pulsed DC as well as AC were tried. Figure 4 shows that when current was applied, in parallel with the load reduction the drawing force was reduced. Reversing polarity caused a smaller electroplastic effect. Pulsed DC is more effective than continuous DC in reducing the drawing force. Figure 5 shows the effects of current density, pulse width and on/off ratio upon drawing force. The reduction in drawing force is seen to scale linearly with current density. These effects are reduced with increased speed of drawing.

Experiments of particular interest in this context consisted of rolling wires fabricated from W, W-Re and Fe-Co--all known as difficult to deform materials--into ribbon under the influence of high-intensity electric current (8-13). Instead of rolling these materials at high temperatures under vacuum, deformation zone temperatures estimated as only 200-300 C sufficed, and superior mechanical properties were obtained. Klimov et al. (8) rolled 0.1 mm W and W-Re wire to reductions in area of greater than 90% using current densities of $10^5$-$10^6$ A/cm$^2$. Troitskii and co-workers (10-14) obtained similar results with W wires. While 0.4 mm wire was rolled into 0.1 x

0.15 mm ribbon without cracking and yielding a product with good tensile strength and ductility, in the absence of current the W wire delaminated and fractured. Klimov et al (12) produced micron thick ribbons of Fe-Co-2% V from 2 mm diameter rod without the intermittent annealing and quenching normally required. In the absence of current, dislocation density was high and cellular structure was largely absent. In the presence of current, dislocation densities were much lower.

### c. Compaction (15,16)

Okazaki (15,16) describes an unconventional but clearly valuable method of compacting powders, even when the powders are oxide coated, without changing their microstructure and properties. Known as electro-discharge compaction (EDC), this method consists of applying a high voltage, high current density pulse (up to 30 kV, $10^6$ A/cm$^2$) for 150-300 $\mu$s to powders already placed under compression. EDC can be used instead of vacuum hot pressing or HIPing, followed by mechanical densification by extrusion, forging or rolling at elevated temperatures. Prolonged high temperature heating is usually involved during conventional processing. In addition to changing the microstructure, oxide formation on the powders almost inevitably occurs. During EDC, on the other hand, the oxide films (both pre-existing and developed during EDC processing) are literally exploded off the particles, reportedly leaving not even discrete oxide particles behind. Densification depends mainly upon input energy rather than upon initial packing density or applied pressure. The speed of EDC and the swift self-quenching of the compact which follows upon termination of EDC prevents degradation of the original microstructure and ensures retention of a fine grain size. These characteristics are particularly important when the powder is produced by rapid solidification processing or by mechanical alloying. However, EDC should also be especially useful when efforts are made to compact powders which readily oxidize, are brittle, or are otherwise difficult to sinter.

More recent work by Okazaki (17-19) has shown that EDC provides a means for consolidating nanocrystalline materials without significant change in grain size. This important result may spur more rapid application of nanocrystalline materials technology.

### d. Recovery (20)

High density pulsed DC ($10^7$ A/cm$^2$, 100 $\mu$s, 2 Hz) accelerated recovery (and recrystallization) in Cu (21), Al (22) and $Ni_3Al$ (23). This is demonstrated by the earlier reduction in the hardness of pulsed specimens relative to those which were not so treated, as shown in Fig. 6. The recovery temperatures were reduced by 50 - 100 C for Cu and 18-35 C for Al, with the amount of temperature reduction diminishing with increasing prior cold work.

### e. Recrystallization and Grain Growth

Low density DC current (up to 3100 A/cm$^2$) had no effect upon grain size of Cu (24) or slightly decreased it (25). Below 550 C, $\alpha$ Ti grain size was decreased by DC or AC current of 1000 A/cm$^2$, but at 600 C, DC current coarsened the grain size (26). Electropulsing is seen in Fig. 7 to have increased the rate of recrystallization of

Cu and $Ni_3Al$. Mainly, pulsed DC current increases by two-fold the pre-exponential, A, in the empirical relationship:

$$(3) \qquad t_{50\%}^{-1} = A \exp(-Q/kT)$$

where $t_{50\%}$ is the time required to decrease hardness by 50% (and by assumption, to recrystallize 50% of the microstructure), Q is the apparent activation energy and kT has its usual meaning. Since the current is on only $2 \times 10^{-4}$ of the total processing annealing time, this increase in A should be regarded as significant. In addition to increasing the rate of recrystallization, electropulsing decreases the as-recrystallized grain size, reduces the frequency of annealing twin formation and sharpens the recrystallization texture (21).

Pulsed DC current had the opposite effect upon the grain growth which followed completion of recrystallization. Grain growth was retarded increasingly as the number of pulses per second was increased, though the retardation was more or less independent of the pulse duration in the range 50-200 μs (22), as shown in Fig. 8. There was some indication that the influence of electropulsing diminished with increasing impurity concentration, though the impurities present and their individual concentrations seem not to have been well defined (22).

### f. Phase Transformations

The pressing industrial problem of electromigration in thin film interconnects has led to use of Al-4% Cu alloys for this purpose. In turn, the effects of DC current upon precipitation in these films have drawn attention, since these precipitates can be a source of voids, and thence failure, at grain boundaries. DC current at densities of about $10^6$ A/cm$^2$ are found to retard significantly $\Theta$ precipitation at both grain boundaries and at the surfaces of thin films (27). Koppenaal and Simcoe (28) found a three-fold increase in reaction kinetics at 75 C, as judged by the variation in electrical resistivity with time, in the presence of DC current densities of 1000-3000 A/cm$^2$. Presumably the precipitate(s) formed did not include $\Theta$; they were not identified in this investigation. AC of 25 and 100 cycles/s at 400 A/cm$^2$ was found to decrease reaction kinetics by a factor of about two.

In 1959, Erdmann-Jesnitser et al. (29) reported that a continuous DC current of 1000 A/cm$^2$ enhanced rates of both quench aging and strain aging of Armco iron at 80 C. However, a continuous AC current of 50 Hz and of the same magnitude as the DC current retarded significantly the kinetics of these aging processes. These effects have been recently confirmed by Campbell and Conrad (30), who found that the critical AC frequency coincides with the jump frequency of the carbon atoms.

Z. H. Lai et al. (31,32) reported that electropulsing ($1.5 \times 10^3$ A/cm$^2$ and 120 μs duration) initiated crystallization in iron-base amorphous alloys at temperatures 90 - 100 C below that at which crystallization begins in the absence of current. The resulting change in microstructure produced a change in the internal magnetic field measured by Mossbauer spectroscopy.

### g. Solidification

Misra (33-35) reported that 30-40 mA/cm$^2$ at 30 V significantly refined the microstructure during the solidification of a Pb-Sb-Sn alloy. Brystkiewicz et al. (36) demonstrated that the growth of epitaxial GaAs crystals from the melt was accelerated in proportion to the electric current (2 - 10 A/cm$^2$) passed through the liquid solution and the substrate. Barnak, Sprecher and Conrad (37) have recently found that electropulsing decreases the grain size of Sn-49% Pb eutectic castings by an order of magnitude. The refinement of grain size by electropulsing occurs for compositions extending from 10% to 90% Sn (38).

## *2. Theory*

### a. Background

The theoretical explanations usually advanced for most of the foregoing effects are a combination of solid state and classical physics. However, in the penultimate paper of the ARO Workshop proceedings upon which this report is based, Kuhlmann-Wilsdorf and Conrad (39) rejected much of this theoretical background and instead ascribed many of these phenomena to "side effects". Their complaints seem well founded, crucial aspects of their objections should be critically tested.

In each of the following subsections, paralleling those in which key experimental observations in each of the areas covered were summarized, the "usual" theoretical explanations will first be presented. The counter-arguments and explanations due to Kuhlmann-Wilsdorf and Conrad, if any, will then be related and discussed; the rebuttal offered by Conrad (40) will then be summarized (together with comments, where appropriate, by the present writers).

### b. Plastic Deformation

Since the 1960's, the view generally taken by physicists is that imposition of an electric current results in interactions of dislocations with electrons and phonons. Anderson (41) emphasizes phonons and phonon/electron interactions, concluding that dislocations and other defects are excited by the current with an energy which is passed to electrons and thence through the phonon "bath" to the environment. However, a variety of thermal resistances can hinder the thermal energy transfers, resulting in heating of the defects. If an electron gas is heated through application of an electric field, the thermal resistance, $R_V$, between the hot electrons and the phonon bath is approximated by:

$$R_V \sim 1/(\beta V T^4) \tag{4}$$

where $\beta$ is the electron density, V is the volume of the sample which is Joule heated, and T is the temperature. Usually, $R_V$ is unimportant for metals, except in very small devices when V is minute. When $\beta$ is small, as in alloys near the metal/insulator transition, metals near the superconducting transition and semiconductors in general, $R_V$ becomes significant.

Interaction of dislocations with thermal phonons can be measured in cryogenic heat-transport experiments. In both ionic and metallic crystals the phonon-dislocation interaction is dynamic. This interaction is described by the "fluttering dislocation" model (42). Heat pulse measurements are accurately predicted by this theory, including the extreme anisotropy of the phonon/dislocation interactions.

Brailsford (43), on the other hand, has emphasized electron/dislocation interactions, such as are more usually considered in analyses of the effects of electric current (and other forces) upon the properties of metals and other materials. A dislocation gliding at moderate velocities is subjected to a viscous drag mainly by the phonons and electrons present. Brailsford had previously reviewed the various contributions of lattice scattering to the drag effect (44) and also the contributions of electronic effects (45). Of these, the electronic effects were evidently considered to be the more important, with further consideration being confined to them.. Using the free electron theory and assuming isotropic continuum elasticity, the following expression was obtained for $B_e$, the electronic drag coefficient:

$$B_e = (1/(2^{3/2}))(c_t/c_l)^2(Gb(\alpha/\omega)) \tag{5}$$

where $c_t$ is the transverse sound velocity, $c_l$ is the longitudinal sound velocity, G is the shear modulus, b is the Burgers vector, $\alpha$ is the attenuation, and $\omega$ is the angular frequency. For a free electron metal similar to Cu, Brailsford (43) obtained $B_e \sim 10^{-5}$ dyn-s/cm$^2$. From the literature, Conrad et al. (2) obtained two relationships for $B_e$. One is derived from considerations of the specific dislocation resistivity (46-49):

$$B_e = \rho_D(en_e)^2/N_D \tag{6}$$

where $\rho_D/N_D$ is the specific dislocation resistivity, e is the electron charge, and $n_e$ is the electron density. The second relationship used by Conrad et al. (2) is based upon quantum mechanical consideration of interactions between conduction electrons and dislocations (50-52):

$$B_e = \alpha b p_F n_e \tag{7}$$

where $\alpha$ is a constant between 0.25 and 1.0, depending upon details of the Fermi surface and the calculations, b is the Burgers vector and $p_F$ is the the Fermi momentum. Conrad et al. (2) obtained $B_e = 10^{-4}$ dyn-s/cm$^2$ from Eq. (6) and $5 \times 10^{-5}$ dyn-s/cm$^2$ from Eq. (7), in adequate agreement with the $10^{-5}$ value Brailsford secured from Eq. (5). Figure 9 (53-56) shows the experimental and calculated variation of $B_e$ with temperature, where "specific resistivity" in this figure represents Eq. (6) and "Fermi momentum" refers to Eq. (7). The experimental data were obtained by Conrad et al. (2,53). Despite the overall similarity of theory and experiment, the latter may indicate a variation of $B_e$ with temperature whereas theory contains no clear temperature dependence in Eqs. (5)-(7). Conrad et al. found similar results for Ag, Al and Cu, but in the case of Nb, $B_e$ was about an order of magnitude higher than predicted. They

suggested that the values of the various parameters in the equations are less well established for this metal.

Mutovin and Klimov (11) also proposed that the influence of an electric current upon deformation arises from an electron push effect upon dislocations. Later, Klimov and Novikov (13) offered another theory, based upon internal temperature and vacancy gradients. The latter theory indicates a dependence upon the second power of the current density. Hence the electroplastic effect should be non-vectorial, in disagreement with experiment.

Kuhlmann-Wilsdorf and Conrad (39) began their complaints about both the experimental results recounted above on the effects of an electric current on plastic deformation by summarizing the results of extensive experiments on electric current effects of electric brushes made of Au-plated Cu wires sliding upon a Au-plated Cu substrate. No effect of high current densities were found. The foregoing effects reported by Troitskii and successors were ascribed to thin oxide films, which should be considerably thinner on Au than on the other metals studied.

A further explanation for the reported effects was proposed to be the following. The magnetic field associated with an electric current must "diffuse" into a conductor. Hence the remainder of the test piece, say a wire, is subjected to a transient mechanical pressure given by:

$$P_{mag} = B^2/2\mu_o, \qquad (8)$$

where B is the magnetic field strength and $\mu_o$ is a constant equal to $4\pi \times 10^{-7}$ Vs/Am. Thus, when B = 1T, $P_{mag}$ = 0.4 MPa. For a superconducting magnet, B = 100 T and $P_{mag}$ = 400 MPa, while the discharge of a capacitor can produce a still higher transient pressure--and is, in fact, used in a successful commercial compactor.

An additional effect results from local heating and leads to large local softening. During machining, for example, Joule heating is concentrated at spots which are already being strongly friction heated. Additionally, since resistivity increases with temperature, these spots can actually melt, providing liquid lubrication where it will do the most good.

As a transient variation on the local heating theme, again because the field cannot instantly penetrate into the interior of the workpiece, Joule heating is initially concentrated at the surface and can accordingly yield much larger temperature increases than anticipated on the basis of the the full workpiece cross-section. In thin wires, surface strength is of especial importance. Hence pulsed DC will be more effective than continuous DC in reducing the force required to produce a given level of deformation, e.g., Fig. 1.

Joule heating is proportional to $j^2\rho$, where $\rho$ is the local resistivity. Hence local high resistivity areas, such as inclusions and grain boundaries, will locally cause further Joule heating.

An additional effect of local heating, particularly on the surface of the workpiece, is thermoelastic stresses. Thus, the 0.2% yield strength level can be reached by 100 C of local heating on the surface of an Al wire.

Attention was then turned to the widely accepted explanation of electric currents upon mechanical properties reviewed above, i.e., the conduction electron push force upon dislocations. However, this force was shown to be too small to account for any of the reported effects, being no larger than 3 MPa. Possibly the phonon drag effect is responsible, since its coefficient can be 10-100X larger than $B_e$.

Magnetostrictive effects at surfaces, on the other hand, are very small.

In the last paper of these symposium proceedings, Conrad (40) attempted a rebuttal of these criticisms by Kuhlmann-Wilsdorf and Conrad (39). "Skin effects", i.e., $P_{mag}$, surface heating, etc. were avoided in the works of Troitskii et al. and Conrad et al. through the use of 1 mm diameter wires, whose radii are less than the calculated maximum penetration depths for electrical and magnetic fields (0.85, 0.66 and 1.0 mm for Al, Ag and brass when the current is either AC or pulsed DC and the application frequency is $10^4$/s). It should be noted, however, that this argument fails to take into account the time dependence of electric and magnetic field penetration into a conducting wire. The "skin effects" will thus be operative until the interior of even a very thin wire has been encompassed by the fields.

Conrad further argues that the polarity effects (5, 57) upon electroplastic behavior are consistent with electron/dislocation interactions in the electron wind sense, as is the dependence of strain rate upon current density. He also notes that the extensively studied phenomenon of electromigration (58) is consistent with the same type of explanation. On the other hand, Conrad notes the aforementioned discrepancies between measured and calculated values of $B_e$ and especially the experimentally observed variation of $B_e$ with temperature and he therefore suggests that perhaps electron/phonon/dislocation interactions will have to be considered, essentially as did Kuhlmann-Wilsdorf (39).

In subsequent work Conrad and Sprecher (53) established that the largest effect of an electric current pulse was on the pre-exponential factor in the thermally activated plastic deformation equation, as illustrated in Fig. 10. This influence on the pre-exponential was attributed by Conrad (59) to an increase in either the entropy of activation or the frequency of vibration of the dislocation as its surmounts the obstacle to its motion.

### c. Compaction

Okazaki (15) concluded that electro-discharge compaction (EDC) occurs in four stages. The first consists of electronic breakdown of the oxide films and heat accumulation at the metal/oxide interfaces. The second is explosive breakdown of the oxide film as a result of sublimation of the metal layer below. The third is neck formation between adjacent particles. And the fourth entails both neck growth and dissipation of energy through the conductive mass. This sequence is consistent with the absence of oxide films and even of oxide particles in the fully compacted structure.

Kuhlmann-Wilsdorf and Conrad (39) add to this scenario, as a corollary to the "skin effect", the peaking of Joule heat at the contact spots between adjacent particles. Local softening and especially local melting will greatly facilitate the compaction process.

### d. Recovery

Sprecher and Conrad (20) ascribe the enhanced kinetics of recovery in the presence of an electric current to more rapid annihilation of dislocations. The more rapid destruction of dislocations is suggested to result from easier cross-slip and dislocation climb, though they cannot explain how current facilitates these processes. However, the well known acceleration of substitutional diffusion by electromigration would explain the quicker dislocation climb. Easier cross-slip, particularly in the presence of pulsed DC or AC current, may result from repeated pressures on dislocations (whether from the electron wind, the $P_{mag}$ effect or any other source) to surmount obstacles hindering their progress. At least in a phenomenological sense, one has the impression that electric currents (as well as electric fields, magnetic, ultrasonic, and microwave fields) tend generally to facilitate the movement of dislocations, whether they are multiplying and moving as during plastic deformation or moving toward destruction as in recovery and to a lesser extent during recrystallization.

### e. Recrystallization and Grain Growth

Conrad et al. (60) suggest that the decrease in recrystallized grain size at relatively low annealing temperatures may arise because faster heating by the Joule effect increases the nucleation rate of new grains. Presumably the reversal of this effect at higher temperatures results from accelerated grain growth following the completion of recrystallization. Michalak and Hibbard (61) found during their studies of recrystallization of OFHC Cu that the prior cold rolling procedure exerted its effects primarily through the pre-exponential term rather than the activation energy for the overall recrystallization process, though again for reasons not yet clear. The acceleration of nucleation kinetics--if such does occur, since there are as yet no data on the effects of electric current on the nucleation and growth kinetics of recrystallization--was ascribed to expediting of the subgrain coalescence mechanism of Hu and Li (62,63). This acceleration might be due to either the influence of electric current upon the vacancy concentration or upon the flux of vacancies to dislocation jogs. The existence of the electromigration effect, however, suggests that reduction in the activation enthalpy for vacancy motion may well be the fundamental reason for faster nucleation, provided of course that nucleation, rather than growth, is the kinetic factor proceeding more rapidly under the influence of electric current.

The explanation offered for a decrease in the rate of grain growth by electropulsing was that it is due to an increased annihilation rate of dislocations (20). However, this explanation refers to the different process of strain-induced grain boundary migration. True grain growth is driven by grain boundary energy reduction, not by dislocation annihilation.

### f. Phase Transformations

Little interpretation was provided of the experimental results on phase transformations summarized in the preceding section. Onodera and Hirano (64) explained their observation on the retardation of aging in Al-4% Cu as a result of electromigration sweeping vacancies into grain boundaries. This explanation is difficult to examine in any detail, however, because of uncertainty as to the phases precipitating and whether their nucleation and/or their growth kinetics were affected by an electric current. In general terms, one might expect that diffusional phase transformations would be expedited primarily because the kinetics of volume and perhaps also grain and interphase boundary diffusion would be increased by the electromigration effect. Martensite transformation kinetics might also be expedited by the readier movement of the glissile dislocations through which both the lattice deformation and the lattice invariant deformation (which together comprise the martensite transformation in non-fcc/hcp transformations) take place. Thus the slowing down of diffusional transformation kinetics is difficult to explain except perhaps by a larger increase in the kinetics of nucleation than of growth (though why this should be affected by an electric current is also unclear). In this circumstance, overlapping of the diffusion fields associated with adjacent precipitates would slow down growth kinetics. Since the influence of growth is far larger than that of nucleation upon overall transformation kinetics, the net effect would be to slow down the overall reaction rates after the initial stages of the transformation. Obviously measurements of nucleation and growth kinetics for an already well characterized diffusional phase transformation under the influence of an electric current is necessary before the atomic mechanisms through which the electric current influences the transformation process can be evaluated. In the case of retardation of quench aging of a low carbon iron by an AC current with a critical frequency, Campbell and Conrad (30) showed that the critical frequency was equal to the jump frequency of carbon atoms.

### g. Solidification

Gupta et al. (65) concluded from their studies on the Cd-Sn eutectic system that a potential source is active within the microstructure of the solidifying system. This was suggested to be either a micro-thermal emf generated by temperature gradients or a potential produced at the solid:liquid interface. The role of these currents in the solidification process, particularly in the presence of a high-density electric current, was not described.

As previously noted, Brystkiewicz et al. (36) found that the epitaxial growth of GaAs from the melt is more rapid in the presence of a low density electric current. Lastrzebski et al. (66) proposed two explanations for this result. One is that Peltier cooling at the substrate:liquid interface increases the undercooling of the liquid, thereby increasing the driving force for growth. The second is that the solute flux toward the advancing solid:liquid interface is increased by electromigration, again increasing growth kinetics. Both explanations yielded good accountings for the experimental results! An attempt to combine them now seems desirable.

Barnak et al. (37) concluded that the reduction in grain size which resulted from the application of high density electric current pulses was due to an increase in the nucleation rate. It was speculated that this may have resulted from a reduction in the

free energy difference between the liquid and solid states or to an increase in the liquid:solid interfacial energy.

## B. External Electric Field Effects

*1. Experimental Observations*

### a. Background

External electric fields as well as direct applications of electrical currents have long been known to cause electromigration (45). Since elevated temperature plastic deformation, recovery, recrystallization, grain growth, phase transformations and solidification all involve point defect movements there is reasonable expectation that these processes will also be affected by external electric fields. As the following subsections will show, this expectation is indeed borne out, but the effects themselves are sometimes the reverse of those resulting from internally applied electric currents.

### b. Plastic Deformation

Conrad and co-workers (2) found that an electric field of 2000 V/cm had little effect upon the stress-strain curve of 1.0 mm thick specimens of either Al or the Al-base alloy 7475 when tested at room temperature. On the other hand, when tested at 520 C, a high homologous temperature, there were significant effects of the same electric field on 7475 as it underwent superplastic deformation. Both the flow stress and the rate of strain hardening decreased and the strain rate hardening exponent increased. Grain boundary cavitation also diminished. Polarity effects were again observed. When the specimen was connected to the positive terminal of the power supply the flow stress required to produce a given strain diminished in proportion to the external electric field gradient whereas connection to the negative terminal increased the flow stress. These effects extended to the center of 1.2 mm thick specimens.

Earlier, Kishkin and Klypin (67) reported that an electric field of 100 V/cm increased the creep rate of Cu and Co about an order of magnitude at high homologous temperatures. Klypin (68) subsequently showed that as little as 0.01 V/cm would slightly increase the creep rate of Cu at 400 C.

J. C. M. Li (69) made ingenious use of the plasto-electric effect to determine the density of mobile dislocations in KCl crystals. Dislocations carry an electric charge in ionic crystals. Since plastic deformation produces a net dislocation flux in one direction a potential difference is simultaneously engendered. This is known as the plasto-electric effect. Knowing the electric charge on a dislocation in an ionic crystal, measurement of the potential difference yields the mobile dislocation density while plastic deformation is in progress. Kataoka and Li (70,71) found that the mobile dislocation density increases during compression at constant strain rate, remains constant during strain rate cycling and decreases to a steady state value during stress relaxation, thereby contributing importantly to our understanding of dislocation plasticity in ionic crystals.

### c. Bonding and Compaction

Although Conrad (49) provided two references to the joining of ceramics, both were found to deal, respectively, with glass-to-ceramic and metal-to-ceramic seals. However, the results may be taken as indicative of the possibility that electric fields may be able to expedite the sintering of ceramics just as electric current (for whatever reason!) markedly accelerated the sintering of metal powders (see the subsection on compaction in the preceding section). Dunn (72) used β-alumina, a superionic conductor, as the ceramic in his experiments. Attempts were made to bond Cu, Kovar, Fe, Mo, Ti, and Al to this ceramic. Voltages applied to produce the electric fields ranged from 50 to 500 V/cm; the resulting currents ranged from 0.5 to 10 mA; temperatures varied from 500 to 600 C; and the current application times were from 45 to 600 min, depending upon the metal being bonded to the ceramic. Bonding was best between Al and β-alumina; the other metals had to be pre-oxidized for bonding to be achieved (which, of course, aluminum normally is!). Surface finish of the ceramic had to be of high quality. Deliberately induced fracture of the Al:β-alumina welds occurred in the ceramic rather than at the interface, as was the case for the other metals bonded to this ceramic.

### d. Recovery (20)

Neither the dielectric environment (vacuum, air, silicone oil) or field strength (in the range 2.4-8.0 kV/cm) exerted a special effect upon the kinetics of recovery of cold worked Al, Cu, α-Ti or $Ni_3Al$. In the range of field strengths applied, the "recovery temperature" (at a given pct. cold work) for Al and Cu increased, i.e., recovery kinetics were diminished. Polarity of the electric field was most important. Only when a specimen was connected to the positive power supply terminal did the external electric field affect the recovery kinetics of Cu. The effects of this field extended to the center of 1 mm diameter specimens. However, the effects of external fields upon the recovery temperature of $Ni_3Al$ were just the opposite of those for Al and Cu, resembling more those of electropulsing, wherein recovery kinetics were accelerated by pulsed DC current. Additionally, no polarity effect was observed in $Ni_3Al$.

### e. Recrystallization and Grain Growth (20)

The effects of an external electric field upon the recrystallization kinetics of Al, Cu, Ti and $Ni_3Al$ reported from this investigation appear to parallel those on recovery.

### f. Phase Transformations (73)

A considerable variety of observations has been presented on the effects of an external field upon phase transformations in steel and in non-ferrous alloys. However, very little of this practically oriented information is sufficiently detailed to allow interpretations to be made of it with much confidence.

For example, Klypin et al. (74,75) reported in 1978-79 that an external electrostatic field of 10-1000 V/cm increased the hardness resulting from heat treatment of Al alloys and a medium carbon steel. In an Al-Cu-Mg alloy, the electric field improved hardness when applied during solution annealing as well as during aging. The influence of the field was larger when this Al alloy was connected to the positive terminal, i.e., when the specimen was the anode. Further, the hardness of the Al-base alloy known as 2024 solution annealed at 500 C for 30 min and then water quenched increases in proportion to the logarithm of the electric field strength. A 7475 Al alloy undergoing superplastic deformation under an external electric field exhibited larger precipitates at grain boundaries and smaller precipitates within matrix grains.

Initial studies indicated that the quench aging of iron was retarded when the specimen was the anode but had no effect when the specimen was the cathode in an external electric field of 10 kV/cm (73). Subsequent work (76, 77) revealed, however, that even though the influence of the field was greater when the specimen was positive, a significant effect also occurred when the specimen was negative. The effects of the electric field included the following: (a) a reduction in peak hardness, (b) elimination of the first of two hardness peaks which occur during aging in the absence of an external electric field, (c) an increase in the apparent activation energy associated with the peak hardness, and (d) a widening of the precipitate free zone wherein intragranular precipitates are absent but grain boundary precipitates grow larger.

In steel the major effect of the electric field occurred during the quench; only a small effect was noted of applying the external electric field only during austenitization. An external field of 1 kV/cm significantly increased the Jominy hardenability of an 0.9% C tool steel, with the influence of the field extending to the center of the 1.6-3.0 mm diameter specimens employed. A 1 kV/cm external field also improved the hardenability of an "02" type steel (whose composition was not given) at intermediate, but not at either slow or fast quenching rates, indicating that the influence of the field was again exerted during the quench rather than during austenitizing. Microstructures appeared not to have been altered by the electric field even though their formation kinetics were changed. The conclusion was reached by Conrad et al. (73) that the main effect of the external electric field was to shift the CCT (continuous cooling transformation) diagram to longer times, with the nose thereof being displaced from ~5s to ~50s. Thus the electric field retarded the diffusional pearlite and bainite reactions. Support for this conclusion was subsequently provided by Conrad's group (78) through their determination of the effect of an electric field on a CCT diagram.

### g. Solidification

No information was found in the subject proceedings on the effects of an external electric field upon solidification kinetics from a liquid matrix. However, Rosenberg (79) noted that an external electric field enhances the crystallization kinetics of amorphous Si, with crystals of Si mapping the field lines.

*2. Theory*

### a. Background

From Gauss' Law, the interior of conductors is free of electric fields other than the minor field associated with electric currents per se (39). Hence any effect of an external electric field upon the interior of a conductor must have been exerted on the workpiece surface and diffused into the inside of the conductor (73). Since sufficient time may not have been available for disturbances on the surface to reach all of the interior regions or the diffusing defect may not be capable of producing the experimentally observed effects ascribed to the external field, the problem of interpreting these effects is thus defined.

### b. Plastic Deformation

In the experiments reported by Sprecher and Conrad (20), summarized in the preceding section, the time required for vacancy diffusion to the specimen center at the test temperatures employed was only 2.5 times greater than that used in the experiment on $Ni_3Al$, but was orders of magnitude larger in the cases of Al and Cu specimens. The diffusion calculation was based upon the simple root mean square (Einstein) penetration equation:

$$s = (2Dt)^{1/2} \quad (9)$$

where s is the penetration distance, D is the vacancy diffusivity and t is the diffusion time. A more accurate formulation, taking into account a concentration gradient of vacancies from surface to center, might have decreased the required diffusion time by up to an order of magnitude. This difference would accommodate the $Ni_3Al$ experiments but not those on Al and Cu.

The Soviet investigator Klypin has been a prolific proposer of theoretical explanations for external electric field effects. Perhaps his most intriguing suggestion is that the polarity effects suggest that a deficiency of electrons reduces the flow stress whereas an excess of electrons increases this stress (75). Another interesting idea is that the changes in creep kinetics described in the experimental observations section results from the interaction of a charged surface layer with crystal defects (68). He also proposed that changes in chemical potential at a specimen surface in the presence of an external electric field will lead to diffusional changes in the chemical potential of phases in the interior (75). Conrad et al. (73) criticized this idea on the ground that the external electric field cannot penetrate the interior of a conductor. However, it seems possible that changes in electron density at the surface due to the external field could be quickly communicated throughout the interior.

With reference to external field effects upon ionic crystals, Li et al. (80,81) have proposed a sweep-up mechanism for the collection of $K^+$ vacancies in their KCl + 88 ppm $Ca^{++}$ alloy by edge dislocations, with thermally activated adsorption and desorption of these vacancies. This theory yielded predictions consistent with experiment.

Conrad (40) noted that in the studies discussed by Li (69), Joule heating was not a factor and electrostrictive stresses were considerably below the yield stress of their material. Kuhlmann-Wilsdorf and Conrad (39) noted that an external electric field gradient of $10^4$ V/cm will generate a friction stress of 2 MPa on dislocations. This would significantly affect dislocation behavior during plastic deformation.

Of particular importance, though, was the suggestion by Kuhlmann-Wilsdorf and Conrad (39) that polarization effects are responsible for the experimental results of Li and co-workers (69). External electric field effects can cause polarization in both metals and non-metals. Substantial attractive forces obtain between the corresponding dipolar surface charges. Since surface charge density and electric field strength increase as the radius of curvature of surface asperities decreases, surface pits and scratches will become the sites of stress peaks. This criticism might be evaluated by repeating the experiments of Li et al. with successively finer surface finishes, and evaluating these finishes with SEM, and if necessary with STM, prior to plastically deforming the same or equivalent specimens.

### c. Phase Transformations

The microstructural changes described in the Experimental Observations part of this section in a 7475 Al-base alloy which had undergone superplastic deformation under an external electric field were explained by the assumption of changes (presumably increases) in the vacancy concentration at the specimen surface. Calculations based on Eq. (9) indicated that these vacancies could diffuse to the specimen center in time to cause the observed effects (73). The present writers elaborate by suggesting that the accelerated growth kinetics of grain boundary allotriomorphs, which the experimental observations clearly imply, result from extra vacancy enhanced volume diffusion of solute to the grain boundaries. On the "collector plate" mechanism (82), this solute diffuses along the grain boundaries to the allotriomorphs. Some attaches directly to the edges of the allotriomorphs, but most of the solute diffuses along the allotriomorph broad faces and then attaches to the allotriomorph faces. The accelerated solute depletion of the matrix attending the more rapid operation of the collector plate mechanism decreases more quickly the supersaturation in the interiors of matrix grains which provides the driving force for the nucleation and growth of intragranular precipitates. Hence the diminished particle number density in the interiors of matrix grains following superplastic deformation in the presence of an external electric field can also be explained on the same basis. If a suitable external electric field similarly affects the formation kinetics of grain boundary $\Theta$ allotriomorphs in an Al-4% Cu alloy, it might be worthwhile to test quantitatively the foregoing explanation by measuring allotriomorph growth kinetics as a function of external electric field. From these data, the volume interdiffusivity in the $\alpha$ Al-Cu matrix would be back-calculated as a function of this field, using the more sophisticated Brailsford-Aaron (83) rather than the original Aaron and Aaronson (82) analysis of collector plate-assisted growth kinetics, and then determining $D_o$ and Q, the pre-exponential constant and the activation energy, for the diffusivities thus obtained. If the activation energy for Al and Cu interdiffusion is diminished by the addition of an external electric field, then evidence will have been provided for an increased vacancy concentration in the presence of an external electric field.

Somewhat similarly, the slowing down of the pearlite and bainite reactions in a steel (whose composition was not provided) was concluded to be inexplicable in terms of surface defects because diffusion to the specimen center was too slow. However, vacancy diffusion is normally irrelevant to diffusional transformations in steel. Except under very unusual circumstances, the growth kinetics of these transformations are controlled by the (interstitial) diffusion of carbon in austenite, as modified by the ledge mechanism (84) and the solute drag-like effect (85). Nucleation may be controlled by alloying element diffusion in austenite grain boundaries (86) but this, too, would be largely or entirely unaffected by changes in the vacancy supply. Only when nucleation is controlled by volume diffusion of a substitutional alloying element in austenite (86) would an enhanced vacancy concentration be important--and yet even then, the resulting effect upon nucleation kinetics would produce no more than a relatively minor increase in the overall kinetics of transformation in view of the much greater importance of growth kinetics (87). At the present time it is unclear how diffusional effects could slow down the kinetics of the pearlite and bainite reactions under "usual" circumstances. However, since both of these reactions take place with an increase in the average volume per atom, if the austenite were to be subjected to a high hydrostatic pressure, or even to high radial compression round the circumference of wire specimens, a diminution of these transformation kinetics could be anticipated on the LeChatelier Principle. On the views of Kuhlmann-Wilsdorf and Conrad (39), an oxide film and/or the $P_{mag}$ skin effect could accomplish the necessary compression. The polarization stress effect, as well as some of the ideas of Klypin, to be discussed shortly, represent other possibilities for explaining these influences upon austenite decomposition kinetics. However, though understanding how an external electric field can slow down the kinetics of diffusional austenite decomposition certainly represents an interesting problem, even if this problem is undertaken on the somewhat simpler proeutectoid ferrite reaction, it appears to be too complicated, at the present state of understanding of both external electric field and alloying elements effects, to be worth attacking at this time.

### c. Grain Growth

Rosenberg (79) reported that $TiSi_2$ precipitation at grain boundaries in polysilicon greatly increases grain growth kinetics. He explains this as the result of $TiSi_2$ serving as a sink for dopant. The resulting flux sets upon a Fermi level gradient in the grain boundary region. This gradient is accompanied by a local electric field which places a significant force on grain boundaries, thereby accelerating their migration. Here an alternate, or perhaps a supplemental explanation is proposed. As in the growth of grain boundary $\Theta$ allotriomorphs at grain boundaries in Al-4% Cu, the kinetics of allotriomorph lengthening and thickening may be presumed to be accelerated by the operation of the collector plate mechanism, described in the preceding subsection on phase transformations. The correctness of this assumption is more likely the smaller the ratio of the activation energy for grain boundary diffusion ($Q_b$) to that for volume diffusion ($Q_v$) is . Thus, the collector plate mechanism is much more important when the matrix has an fcc crystal structure (88) than when its crystal structure is bcc (89). For self-diffusion, $Q_b/Q_v$ ~ 0.5 for fcc metals and ~ 0.6 for bcc metals (90). For grain boundary interdiffusion in Si, $Q_b/Q_v \sim 0.7 - 0.8$ (91); no data were found for grain boundary diffusion of Si in Si (91). While the component of diffusion along the grain boundaries of the collector plate mechanism reproduces the

diffusion path taken during diffusion-induced grain boundary migration (DIGM), the high ratio of the Q's for the two types of diffusion indicates that $TiSi_2$ precipitation at grain boundaries may significantly accelerate grain growth only at relatively low temperatures by means of the DIGM mechanism. This suggestion could be tested in a simple way by using analytic electron microscopy to ascertain whether or not grain growth is accompanied by extensive Ti partition only in regions where grain boundary migration has occurred. Clearly, this test would be effective only if performed during early stages of grain growth.

## C. Magnetic Field Effects

*1. Experimental Observations*

### a. Introduction

Hochman and Tselesin (92) note that as of the date of this workshop (1989), there are more than 500 references on (usually pulsed) magnetic field effects upon the structure and properties of alloys. Most of these papers and books were published in the former USSR. Unfortunately, their paper on magnetic field effects (the principal source of information on these effects in the workshop proceedings) is considerably better at naming processes upon which magnetic field effects were generated than upon saying what was learned about these effects! Hence, while magnetic field effects may well be as important as those of electric currents and external magnetic fields, the following subsections, and especially the subsection on Theory, will likely not convey this impression to the reader.

As an illustration of this problem, the first identified worker in this field is Alekseev, who published in 1937 in an unidentified journal in the USSR, and found that magnetic treatment increases the hardness of HHS steel. The chemistry of this steel is not given. Further, there is no indication as to whether the hardenability of this steel was increased (so as to ensure that it transformed to a lower temperature reaction product--say martensite), the internal structure of the transformation product was somehow changed or some other effect was involved. More broadly, Hochman and Tselesin cite monographs (93-96) and several major reviews (97-100) in which magnetic treatments of both ferromagnetic and non-ferromagnetic (101,102) materials are reported. Magnetic field effects are noted on "physical-mechanical" properties of treated materials, diffusion and transformation processes, residual stress and stress relief, wear resistance and fatigue properties. Unfortunately, elaboration on these interesting topics in the following subsections will prove less complete than either the reader or the writers would wish.

### b. Deformation

During electron transitions in a magnetic field, changes were found in magnetic, thermal, optical and mechanical properties (93,102). These include increased hardness. Ferromagnetic materials exposed to a pulsed magnetic field experienced microscopic deformation, leading to magnetostriction substructural strengthening. For paramagnetic Nb and Mo, a constant magnetic field of 1-1.5 KOe caused the yield point to drop faster with increasing temperature, increased stress

relaxation at constant strain, increased plasticity within certain temperature ranges, caused a small decrease in the Peierls activation energy and increased the mobility of screw dislocations (77).

Cadek et al. (103) found that the apparent activation energy for steady state creep of carburized Armco iron diminishes abruptly at the Curie temperature (1042 K). At higher temperatures, the apparent activation energy depends (anomalously) upon both temperature and stress.

The S-N fatigue curve was raised to higher stress levels after quenching and tempering one steel at 250 C and normalizing a second steel from 940 C and annealing at 660 C. A monograph by Malygin and Varulenro (98) was said to give the only conclusive results on this subject.

Cullity et al. (104) found a dramatic increase in stress relaxation in a Ni steel deformed in compression. Very low concentrations of Fe, Cr and Mn in commercial materials provide localized magnetic moments resulting in unique (but unstated) changes of properties when subjected to a magnetic field (105-108).

Extensive Soviet research on the influence of magnetic fields upon wear resistance and cutting tool life has been reported, leading to the following results (94):

(i) Wear resistance was increased by a factor of up to 2.5 on 18% W and on 6% W-5% Co tool steels following magnetic treatment;

(ii) Tool life increases or decreases, depending upon polarity of the magnetic field in the cutting tool and the direction of the feed;

(iii) Stronger magnetic fields can increase high-speed-steel tool life by 1.5-3X. Beyond a certain limit, however, the tool bit can be embrittled and the cutting edge destroyed;

(iv) Multiple remagnetization of a cutting tool in a relatively weak field (5-5.5 Oe) by rotating the tool between different poles of an electromagnet at constant current increases tool life;

(v) Changes occur in the coefficient of friction between a magnetically treated cutting tool and the workpiece.

Hochman and Tselesin (92) have used positron annihilation spectroscopy to show that recovery following cold work is quickly accelerated by application of a pulsed magnetic field. After many reversals, there appears to be a change in the dominant trapping state. Al'shitz et al. (109-111) then reported the discovery of a magnetoplastic effect involving a considerable increase in the plasticity of both metallic and ionic crystals upon application of a magnetic field in which $B \leq 2T$.

### c. Phase Transformations

J. W. Morris (112) has reported (only in an abstract; he was unable to participate in the workshop and did not submit a paper to the proceedings) that

magnetic fields exert significant effects (presumably in steel) only in metastable austenite which can be transformed to martensite during plastic deformation. The effect of a magnetic field is to influence the proportion of the austenite which is transformed to martensite, though it was not made clear whether or not the proportion of austenite increased or diminished. No mention was made of possible polarity effects.

Hochman and Tselesin (92) report that magnetic fields influence the tempering of martensite and the precipitation of carbides. The wording used suggests that tempering may be accelerated. The phase from which the carbides precipitate is not given nor is the type of kinetic change indicated.

*2. Theory*

### a. Introduction

In general terms, Soviet investigators (93,94) envisage magnetic treatments as making changes in the properties of metals through a combination of effects. These include: (i) irreversible changes in structure and properties (neither of which is specified) of both ferromagnetic and non-ferromagnetic metals; (ii) reorientation of the magnetic moment of single defects and of combinations of defects, which becomes equivalent to applying an additional external stress; and (iii) despite the inability of the magnetic fields usually applied to create new defects, the energy thereby supplied can be sufficient "to initiate some processes driven by the inner energy of the distorted crystallographic lattice".

### b. Deformation

Theoretical studies have been reported upon both the effects of dislocations upon magnetic properties and the effects of ferromagnetism upon dislocation movement. Here we shall consider mainly the latter. Vicena (113) made one of the first theoretical studies of dislocation/ferromagnetic domain wall interactions; though oriented toward magnetic properties effects, this work has proved equally useful in the reverse direction. Scherpereel, Kazmerski and Allen (114) extended the magnetoelastic calculations of Vicena to both 180° and non-180° Bloch walls and to edge, screw and mixed dislocations in Fe and Ni, using the conceptual framework described by Kittel (115). Scherpereel et al. calculated the magnetoelastic energy, viewing it as any change in the magnetic anisotropy energy density resulting from elastic distortion by the dislocation of the ferromagnetic lattice. An anisotropy energy density function, $f_k$, is expanded in a Taylor series in the components of strain, yielding:

$$f_k = (f_k)_o + \Delta f_k{}^M + f_{me} \qquad (10)$$

where $(f_k)_o$ is the anisotropy energy density of the initial lattice, undistorted even by magnetostriction, $\Delta f_k{}^M$ is the magnetoelastic energy density due to spontaneous magnetostriction and $f_{me}$ is the magnetoelastic energy density associated with lattice distortions other than magnetostriction, which can be written as summations involving two magnetoelastic coupling constants, direction cosines of the local magnetization

with respect to the cube axes and components of the symmetric strain tensor. The magnetoelastic energy density is then integrated over the distance between the dislocation and the nearest domain wall, as well as through the (calculated) thickness of the wall. The calculations were performed as a function of the several variables involved for both Fe and Ni, yielding quite different results. Overall, however, the interaction energies are very small, corresponding at room temperature to about 4kT per atomic distance along the dislocation. Hence the effect of magnetic changes on the glide of individual dislocations is small (as shown by previous investigators). However, because magnetoelastic effects are additive, arrays of relative immobile dislocations in subboundaries developed during deformation can markedly affect defect structure-sensitive magnetic properties, e.g., the coercivity of Ni can be increased 20-30X by cold work. This result can be interpreted in terms of a domain wall which must be driven through material with a large and locally varying density of dislocations.

Kuhlmann-Wilsdorf and Conrad (39) point out that the electronic drag on dislocations is also too small to explain the observed effects of an externally imposed magnetic field. As discussed in the section on the theory of electric current effects, a magnetic field also cannot penetrate a metal instantly. At steady state the penetration depth is only about $10/(\nu)^{1/2}$, where $\nu$ is the the frequency of application of the magnetic field. However, this penetration can briefly exert a high $P_{mag}$ upon the workpiece (see Eq. 8), as in the case of directly applied high-density electric currents. Magnetostrictive stresses are also too small to yield significant strength changes, but surface film-based effects could be triggered by these stresses. The point was also made that magnetostrictive effects upon testing equipment--which tends to be both larger and more rigid than the specimens being tested--should be taken into account.

Molotskii (116,117) has proposed that the magnetoplastic effect results from electronic intercombination transitions between radical pairs and triplet states associated with paramagnetic impurities.

### c. Phase Transformations

Although no theoretical considerations were provided about magnetic field effects upon phase transformations, the ability of these effects to encourage the formation of martensite (112) suggests that perhaps the small interaction energy between magnetic domain walls and dislocations may be sufficient to trigger the growth of a dislocation-swathed martensite embryo. This action could occur by accelerating the dislocation rearrangement process through which martensite embryos develop on the Olson-Cohen mechanism (118-120) or perhaps by initiating "prematurely" the growth of a martensite embryo thus developed. The compressive stresses normally exerted by the $P_{mag}$ skin effect would appear to inhibit martensite nucleation in steel, where this transformation takes place with an increase in the average volume per atom.

## D. Microwave Effects

### *1. Experimental Observations*

#### a. Introduction

The only experiments reported on microwave research in the Workshop proceedings dealt with effects upon processes taking place in ceramics.

#### b. Sintering of Ceramics (121)

Since microwaves can penetrate most ceramics, uniform heating through the volume of ceramic particles is feasible. This reduces temperature gradients and thus thermal stresses, to which ceramics are particularly vulnerable because of their inherent brittleness. Advantage has been taken of this characteristic in experiments on the sintering of alumina powder. With 28 GHz microwave radiation, alumina could be densified at temperatures 300 - 400 C lower than in the absence of such irradiation. A uniform, fine grained microstructure resulted. Density of the microstructure increases faster in microwave than during conventional sintering. The apparent activation energy for sintering is reduced from 575 kJ/mole to 170 kJ/mol when microwave radiation is employed.

#### c. Grain Growth in Ceramics (121)

Grain growth in alumina at 1500 C under microwave irradiation was found to be faster than conventional grain growth in alumina at 1625 C and about as fast as during conventional grain growth at 1700 C. The apparent activation energy for grain growth under microwave irradiation was 480 kJ/mole whereas that for conventional grain growth was 630 kJ/mole.

### *2. Theory*

Kimrey and Janney (121) suggested that the reduction in the apparent activation energy for sintering effected by microwave irradiation might be due to enhancement of aluminum and oxygen ion diffusion. However, the much higher apparent activation energy for grain growth is puzzling in this context. Sintering can successively involve surface, grain boundary and volume diffusion as rate controlling processes, whereas grain boundary migration--in the absence of impurity effects--should depend only upon trans-grain boundary diffusion. Hence one might expect lower activation energies for grain growth than for sintering unless the activation energy for the latter process is evaluated only at an early stage. In the presence of impurities, the activation energy for grain growth can rise toward that for volume diffusion, the process by which impurity atoms are dragged along by the grain boundaries to which they are bound. However, similar effects should be exerted upon sintering if the alumina powder and the atmosphere used are the same for the sintering and the grain growth experiments. One may tentatively conclude that the apparent activation energies reported are an unreliable tool for assessment of the atomic mechanism through which microwave irradiation expedites sintering and grain growth in ceramics. A full theoretical expression for the rate of grain growth in the

presence of impurities and a counterpart equation for sintering kinetics at the stage being experimentally investigated are prerequisites to analysis of the rate controlling step in these processes.

Kuhlmann-Wilsdorf and Conrad (39) point out that the "skin effect" and local Joule heating at contact spots will also sharply raise the local temperature during exposure to microwaves. This effect can be significant during microwave heating of ceramics because of their low thermal conductivities. Temperature peaks at grain boundaries should also be considered during microwaving of ceramics.

The kinetics of the acoustoplastic effect are considered in a paper by Kozlov and Selitser. Kozlov et al. (122) subsequently reported on the effects of ultrasonic vibrations on the plastic deformation of metals (123).

## E. Ultrasonic Effects

### *1. Experimental Observations*

#### a. Introduction

R. E. Green (124) notes that high-intensity ultrasound (HIUS) has found practical applications in welding, machining, drawing, forming, grain refinement during solidification, diffusion enhancement, accelerated fatigue testing and stress relieving. Although Conrad (31) states that HIUS usage in metal working and processing was active about 35 years ago (from the present date), this interest soon disappeared; he calls attention to results of the type now to be described and urges that HIUS effects be reconsidered.

#### b. Deformation

Green (124) summarizes the principal effects of HIUS as work softening, work hardening, and reduction of fracture of tools and their workpieces. Langenecker and co-workers (125,126) reported that the shear stress required for plastic deformation is reduced by HIUS in Zn, Al, Cu, and steel. The reduction in tensile stress during deformation of Zn single crystals amounted to 40%, but promptly disappeared when HIUS was no longer applied. Of especial military and industrial interest is the work of S. P. Kundas et al. (127) on the flattening of W and Mo wires heated in vacuo with 18-44 kHz, 0.4-4.5 KV energy HIUS irradiation. These wires were successfully flattened with a 75% deformation, without cracking and with satisfactory ductility remaining. Without HIUS processing, however, W wire delaminated and broke during the deformation used to flatten it. Thoriated W wire is used to reinforce superalloys (128) and possibly also in penetrators. As presently available, however, this wire is seriously cracked during processing and these cracks are an important source of failure in jet engine turbine blades made from a wire-reinforced superalloy (128).

In an earlier review (129), Green provided a good summary of experimental observations on HIUS effects upon softening of deformed metals. The amount of stress reduction achieved was found to be directly proportional to HIUS intensity but independent of frequency in the range 1.5Hz to 1.5MHz. More thermal energy than

ultrasonic energy was needed to produce the same stress reduction. This effect was found to be independent of temperature from 30 to 500 C.

Localized heating under HIUS irradiation was found to occur at attachment points to specimens, displacement nodes, saw-cuts, drilled holes, fatigue cracks and grain boundaries, as demonstrated by infra-red thermography (124). X-ray diffraction topography performed with a synchrotron showed that HIUS caused severe plastic deformation and lattice bending through the volume of the mid-section of Al specimens.

### c. Role of Dislocations in Hydrogen Embrittlement (130)

HIUS was used to study the role of dislocations in the hydrogen embrittlement of Group VA metals with hydrogen. Ultrasound of 20KHz with a strain amplitude of $10^{-4}$ was superimposed on static loading to study ductility of hydrogenated Nb between 78 and 373 K.

In the absence of hydrogen, HIUS had no effect above 150 K and little below that temperature. However, in the presence of dissolved hydrogen, HIUS considerably reduced the yield stress at all temperatures, especially at lower temperatures when the presence of niobium hydride is expected. This presence led to more reduction in the yield stress than would otherwise have occurred. (Unfortunately, the assumption that hydride was present was evidently not checked with TEM.) Increasing the concentration of hydrogen from 70 to 170 ppm halved the effect of HIUS. Ductility was reduced in the presence of hydrogen and the ductile-brittle transition temperature was increased by about 50 K.

### d. Recrystallization and Grain Growth

Recrystallized Mo strip which had been rolled under the influence of HIUS vibrations had a much finer grain size after recrystallization than conventionally cold rolled strip. However, it was not clear whether the finer grain size resulted from higher nucleation and/or slower growth kinetics. In view of the greater ease of slip in acoustically irradiated material, low rates of growth of recrystallized into unrecrystallized grains seem possible. On the other hand, readier dislocation movement may also make development of subgrains into sizes large enough with dislocation densities sufficiently small so that they can serve as "nuclei" for recrystallization more readily than conventionally processed strip. Hence a serious investigation into the nucleation and growth kinetics of recrystallization under the influence of HIUS (and other) fields would be of considerable fundamental as well as practical interest.

### e. Diffusion

Hochman (131) examined the effect of HIUS irradiation upon the interdiffusion of Hg into $Hg_3Sn$. Although diffusion was accelerated, the formation of $Ag_2Hg_3$ and $Sn_8Hg$ (the "$Hg_3Sn$" was actually a dental amalgam in which other elements were present) complicated the analysis to the point where the results seem to have little value. Ultrasonic irradiation at elevated temperatures would undoubtedly

pose significant experimental problems, for example in coupling the transducer to the specimen. However, using high purity materials and designing the experiments to involve only binary interdiffusion in a single phase field would greatly ease the problem of interpretation. If possible, HIUS effects upon radiotracer self-diffusion ought to be studied first, using a pure metal in which the values of $D_o$ and Q in the absence of irradiation are well accepted and in which the melting temperature is low enough so that volume diffusion will readily occur in an experimentally convenient temperature range.

#### f. Sintering

Hochman (131) studied the influence of HIUS vibrations upon the sintering of Pb, Mg, Zn, Sn and Al powder compacts. Powders were sintered at room temperature while under the high compressive pressure with which they were originally compacted. Products of this procedure had a higher density and a higher compressive strength when extruded into wire than those produced by conventional sintering. Additionally, HIUS-sintered Pb had a higher tensile strength than that conventionally prepared.

### *2. Theory*

#### a. Introduction

Kuhlmann-Wilsdorf and Conrad (39) did not level their litany of serious questions and complaints against the experimental results and theoretical explanations of the influence of high intensity ultrasonic irradiation upon metal properties. However, their views seem applicable to at least some of the findings and explanations noted in this section. Hence an effort will be made to transfer certain of their critiques to the present situation.

#### b. Deformation

Blaha and Langenecker (125) have suggested that HIUS irradiation leads to heating in the immediate vicinity of dislocations and also drastic changes in Young's modulus. Green (124) considers that HIUS vibrations do cause dislocations to move, to multiply, and to interact with other dislocations. He also noted that when a stress, either continuous or pulsed, greater than a threshold level is applied, marked decreases occur in the effective second order elastic moduli. Further, non-linear effects cause elastic moduli to decrease even in the absence of defects. The presence of dislocations causes a still greater non-linear decrease in elastic moduli. These ideas suggest that if measurements of elastic moduli can be made in the presence of HIUS vibrations, a critical test of them could be performed. However, the usual method of making such measurements does involve high frequency vibrations. Hence a different, perhaps less direct approach would have to be taken to perform such experiments.

From the viewpoint of Kuhlmann-Wilsdorf and Conrad (39), a question must be raised as to the extent of local heating associated with HIUS vibrations. The aforementioned non-linear effects upon elastic moduli aside, the more intense

vibrations expected at and in the immediate vicinity of dislocations (because the atoms in such regions are displaced from their normal minimum energy positions) may raise the local temperature enough to reduce the applicable elastic moduli sufficiently to account directly for the greater ease of plastic deformation in the presence of HIUS irradiation. Also on the Kuhlmann-Wilsdorf-Conrad approach, concern should be expressed as to the ability of HIUS to "nucleate" dislocations. The atomic displacements required to accomplish this seem excessive relative to the likely amplitudes imposed by HIUS irradiation.

### c. Hydrogen Embrittlement (130)

Hydride precipitation from Nb leads to punched out dislocation loops as a result of the large volume change involved (132). These dislocations facilitate plastic deformation and thus reduce the stress required to introduce sufficient mobile dislocations to produce yielding. The halving of the effect of HIUS in reducing the yield stress when the hydrogen concentration is increased from 70 to 170 ppm is explained by the increase in the number of dislocations which can be shaken free of hydrogen atoms when the hydrogen concentration is smaller. On the other hand, the decrease in ductility at a given hydrogen concentration with decreasing temperature, even in the presence of HIUS irradiation, is suggested to result from accelerated hydrogen diffusion along dislocations to crack tips (133-135). The increase in the number of mobile dislocations resulting from HIUS application will increase the number of "short circuit" conduits feeding hydrogen to the crack tips, thereby encouraging continued growth of the cracks.

### d. Sintering

The accelerated sintering found in the presence of HIUS irradiation may be due in part to softening and even melting at the spots where adjacent particles are compressed into close contact with one another. The circumstance that the sintering experiments under HIUS were conducted while the particles were under high levels of compressive stress and at relatively high temperatures compared with that of their melting temperature makes this a plausible possibility for enhancing the kinetics of sintering. If alloy particles were to be substituted for high-purity metals, choosing alloying elements which form a eutectic with the solvent metal, TEM study of the sintered compact could disclose evidence for melting in the form of eutectic microstructures in regions where particles were compressed against their neighbors.

## III. FORMING OF COMPONENTS MADE OF HARD-TO-FORM MATERIALS

Advantage can be taken of some of the most significant effects described above (Section II) to solve several problems associated with the manufacturing of many low ductility material components for military and civilian applications. Salient among these effects are those caused by high density electric fields, electric currents, and ultrasonic fields, applied singly or in combinations. Following the lead of Troiskii et al. (138, 139), pulsed current, followed by high-intensity ultrasonic irradiation (HIUS), may be used in addition to the application of only one of these high-field stimulants during (or just prior to) the application of the mechanical load from the forming operation. High-intensity electric fields may similarly be added to high-intensity ultrasonic irradiation as well as also being separately employed. Koslov et al. (123) have

reported that "electroplastic" and "acoustoplastic" effects are additive. In addition to checking their findings, an effort should be made to apply *simultaneously* HIUS and a high-intensity electric field.

A wide range of military and civilian applications could be significantly impacted by forming components to near-net-shape with the aid of high fields. They include those using materials that heretofore have defied forming and others which require extensive, and therefore expensive, machining of castings of difficult-to-machine materials. It should be noted that, in either case, forming with the aid of high fields maintains or improves the mechanical properties of materials processed without their aid. Preeminent among these materials are: the titanium aluminide alloys which may be used to manufacture wrought gas turbine blades, carbon seal support rings, and other high temperature engine components; the tungsten alloys which may be used in missile nozzles, kinetic energy penetrators for gatling guns, shaped charge liners, etc.; and some metal alloys such as Alloy 718, Ti-6Al-4V, and Fe-Co (in monolithic or powder form) used in a variety of DoD applications.

The microstructure of near-$\gamma$ TiAl alloys is complex. The number of papers published in this area, and especially upon the effects of microstructure on mechanical properties, has become extraordinarily large in recent years. A lamellar structure consisting mainly of thin $\gamma$ TiAl lamellae, interspersed with less than ~20% of still thinner $\alpha_2$ $Ti_3Al$ lamellae appears to provide considerably better ductility--though still only a few pct. at room temperature--than either 100% $\gamma$ or $\alpha_2$ (140). However, a microstructure in which $\gamma$ is largely composed of approximately equi-axed crystals and $\alpha_2$ is largely confined to precipitation along $\gamma$ grain boundaries has been found to have improved, though still limited ductility in Ti-47.5% Al-2.5% Cr (141). This type of microstructure yielded similar results in a Ti-47% Al-1% Cr-1% V-2.5% Nb alloy, but unfortunately also inferior fracture toughness (142). A Ti-49% Al-3.4% Nb-0.07% Si alloy also produced poorer fracture toughness in conjunction with both $\alpha_2$ and $Nb_5Si_3$ precipitation at $\gamma$ grain boundaries (143). The origin of the equi-axed $\gamma$, though not much discussed in any of the referenced papers, is probably the recrystallization of this phase and possibly also of $\alpha_2$ under the driving force of the cold work introduced during processing of the alloys. The drastic changes in polyphase microstructure resulting from recrystallization were first recognized and explained by C. S. Smith (144, 145). Since ordered hexagonal $\alpha_2$ is much less plastically deformable on multiple slip planes than is ordered face centered cubic (actually slightly tetragonal) $\gamma$ (146), it is not surprising that extensive formation of $\alpha_2$ along grain boundaries leads to inferior fracture toughness. However, it appears that the closely spaced lamellar boundaries are particularly efficient crack stoppers; hence the lamellar structure provides greater fracture toughness, though at the expense of less ductility than the recrystallized (usually termed "duplex") microstructures.

Trace element additives which increase the ratio of the $\alpha_2$:$\gamma$ boundary energy relative to that of $\gamma$:$\gamma$ grain boundaries in order to limit the spreading of $\alpha_2$ along $\gamma$:$\gamma$ grain boundaries might be considered in order to preserve the superior ductility of

grain boundaries might be considered in order to preserve the superior ductility of microstructures based upon equi-axed $\gamma$ grains while reducing the deleterious effects of $\alpha_2$ upon fracture toughness. Luster and Morris (141) have recently explained this difference in ductility in terms of the far greater geometric difficulty of transmitting slip across lamellar interphase boundaries than across random boundaries. Some of the random boundaries, they point out, are likely to make such transmission occur readily as a consequence of the more favorable geometry of the crystals comprising them.

The effects of HIUS and high-density electric current or of HIUS and high-intensity electric fields, applied either singly or together, should thus be examined for a variety of microstructures, beginning with alloys capable of producing 100% $\alpha_2$ and 100% $\gamma$, respectively, and then moving on to representative $\alpha_2 + \gamma$ lamellar and equi-axed microstructures. Improvements in ductility would be of especial interest from the standpoint of formability. A finding that the formability of one phase is improved more than that of the other by high field effects could induce a change in the type of microstructure emphasized. However, if the most readily formable microstructure does not yield an acceptable combination of strength, ductility and toughness in the as-formed condition, consideration should be given to either a post-deformation heat treatment or to a modification of the microstructure prior to field-aided deformation such that a reasonable compromise can be achieved between the requirements of deformation processing and those of service in a gas turbine engine.

As described in Section II, the rolling of W, thoriated W, W-Re and Fe-Co alloys, which is quite difficult using conventional procedures, is much facilitated by high-intensity pulsed DC current (8-13). The Fe-Co alloy studied was presumably an ordered bcc intermetallic compound. High-intensity electric fields were found to facilitate the high temperature deformation of Cu, Co and both Al and an Al-base commercial alloy (2,67,68). An external electric field has been shown to accelerate the recrystallization kinetics of $Ni_3Al$ (20), suggesting that dislocation movement is also facilitated by this type of field. Similarly, high-intensity ultrasonic irradiation markedly facilitates the rolling of Zn, Al, Cu and steel (124-126) and the flattening of W and Mo wires (127). Not only are less force and lower temperatures needed to accomplish the required deformation but also the quality of the product is much improved, particularly from the standpoint of freedom from cracks (127).

Finally, it should be pointed out that preliminary estimates indicate that forming with the aid of high fields may be accomplished in components up to approximately 20 inches in diameter and one inch thick with commercially available power sources. Moreover, the cost of the laboratory equipment needed for its development and the capital equipment required for production fall within the respective costs of equipment used for similar processing operations.

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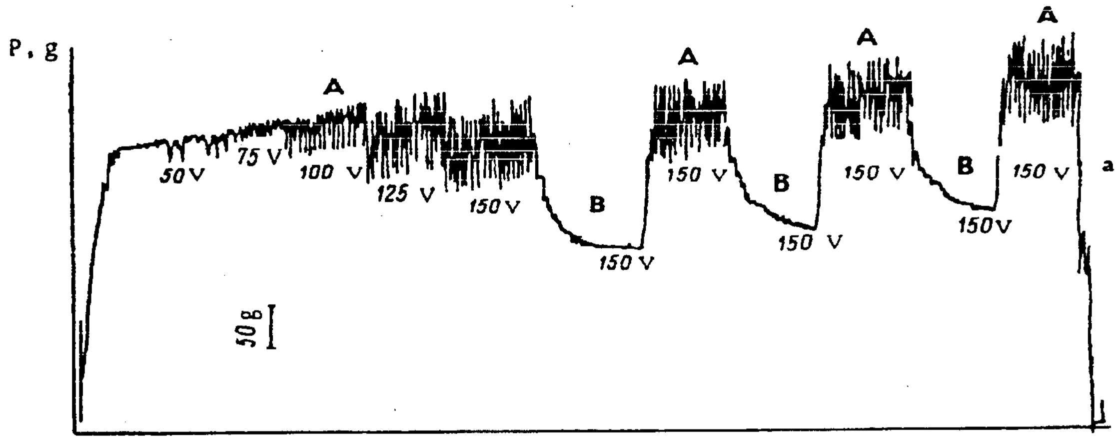


Fig. 1 - Load vs. extension diagram for a zinc crystal in uniaxial tension at 78K with a strain rate of $1.1x10^{-4}$ $s^{-1}$ showing load drops resulting from the application of d.c. pulses produced by discharging capacitors with the voltages indicated (100V - 1.5 x $10^5$ A/cm$^2$. Regions A correspond to constant extension rate. Regions B to stress relaxation (i.e., the test machine was off). From Troitskii (1).

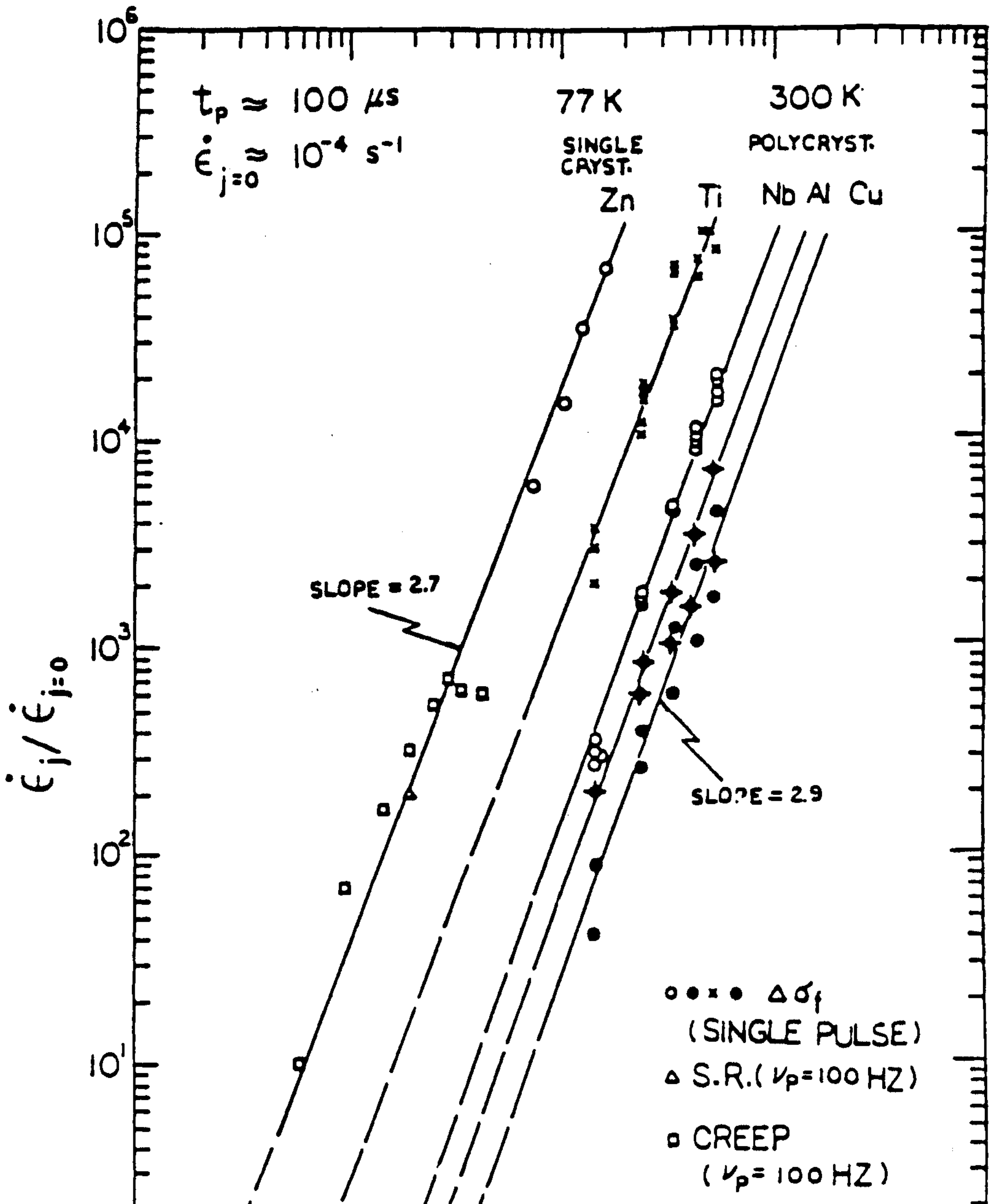


Fig. 2 - Log-log plot of the ratio of the plastic strain rate $\dot{\epsilon}_j$ produced by an electric current pulse to the applied strain rate $\dot{\epsilon}_{j=0}$ prior to the pulse for Zn single crystals at 78K and a number of polycrystalline metals at a 300K. Test methods include constant strain rate test to yield the drop in flow stress for a single pulse $\Delta\sigma_f$, stress relaxation S.R. and creep tests with 100 pulses per second.

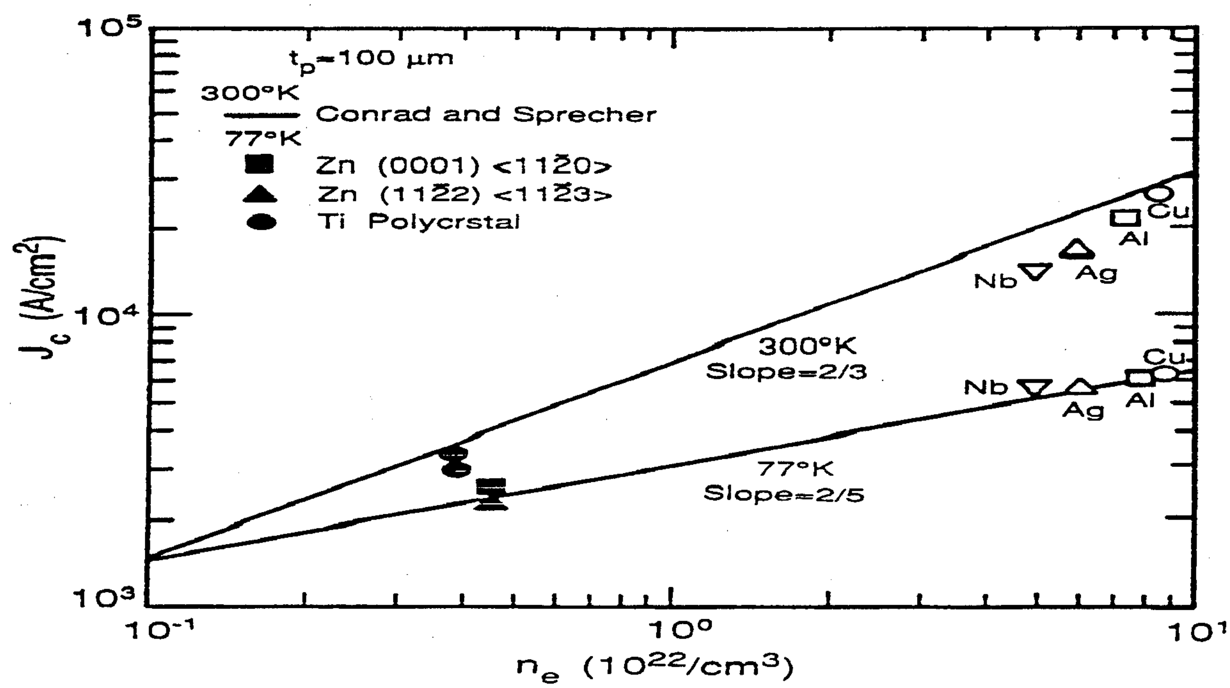


Fig. 3 -Log-log plot of the critical current density $j_g$ at 77 and 300 K vs the electron density $n_e$.

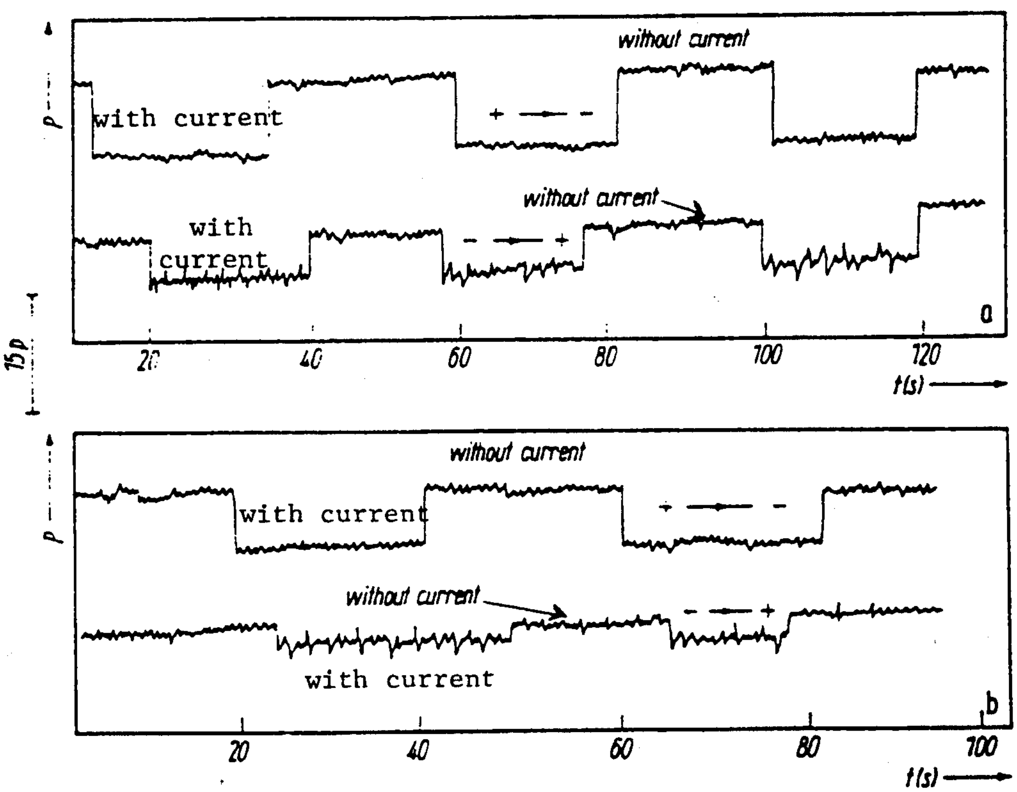


Fig. 4 - Typical diagrams of the drawing force reduction of copper wire 60 μm in diameter with and without the application of an electric current to the deformation zone: mean effective current density $J_{mean.eff} = 3.5 \times 10^5$ A/cm²; wire movement rate = 0.45 m/s; reduction ration = 16.1%: (a) pulsed current with a frequency of 10 kHz and a pulse duration of 60 μs; (b) direct current with fluctuation not exceeding 10%.

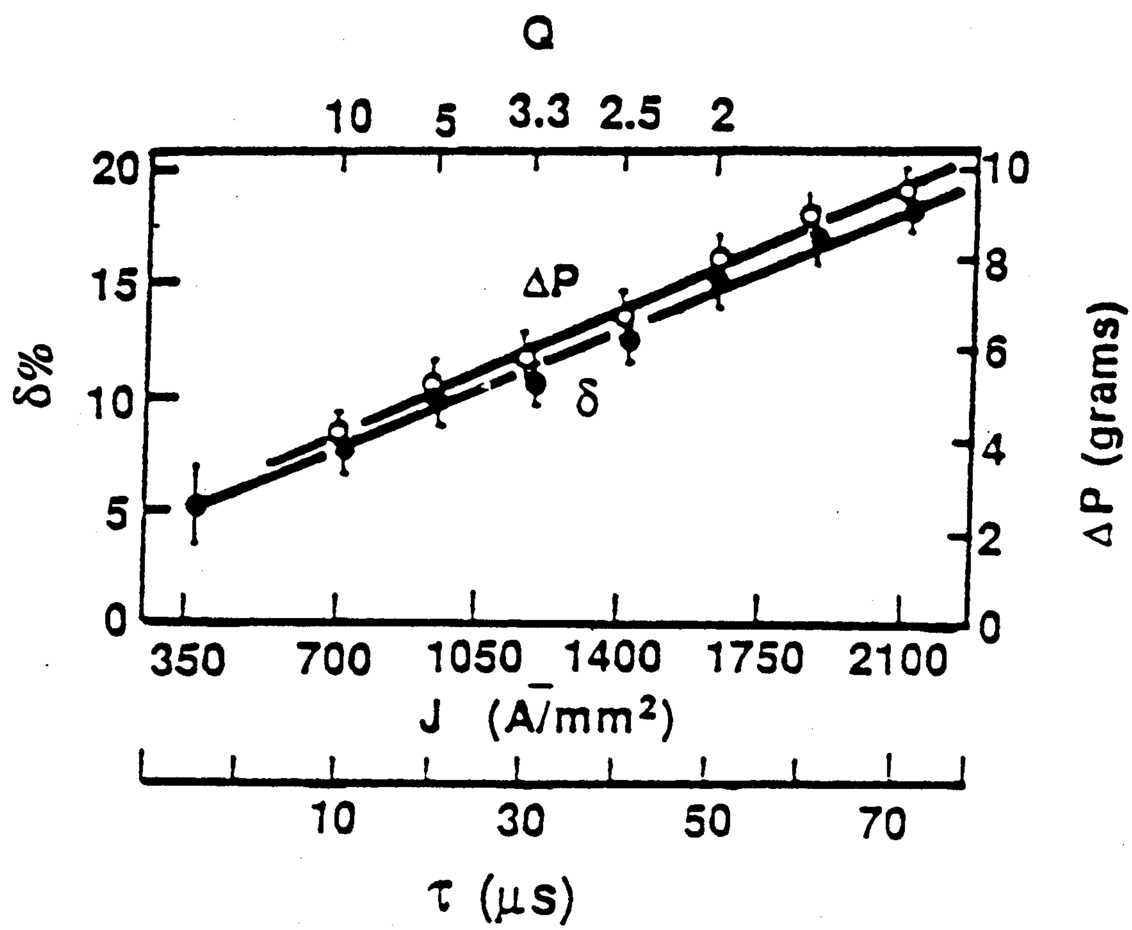


Fig. 5 - Dependence of the absolute load drop, $\Delta P$, and the relative load drop, $\delta = (\Delta P/P)$ 100% versus current density (J), pulse duration ($\tau$), and on-off ration (Q).

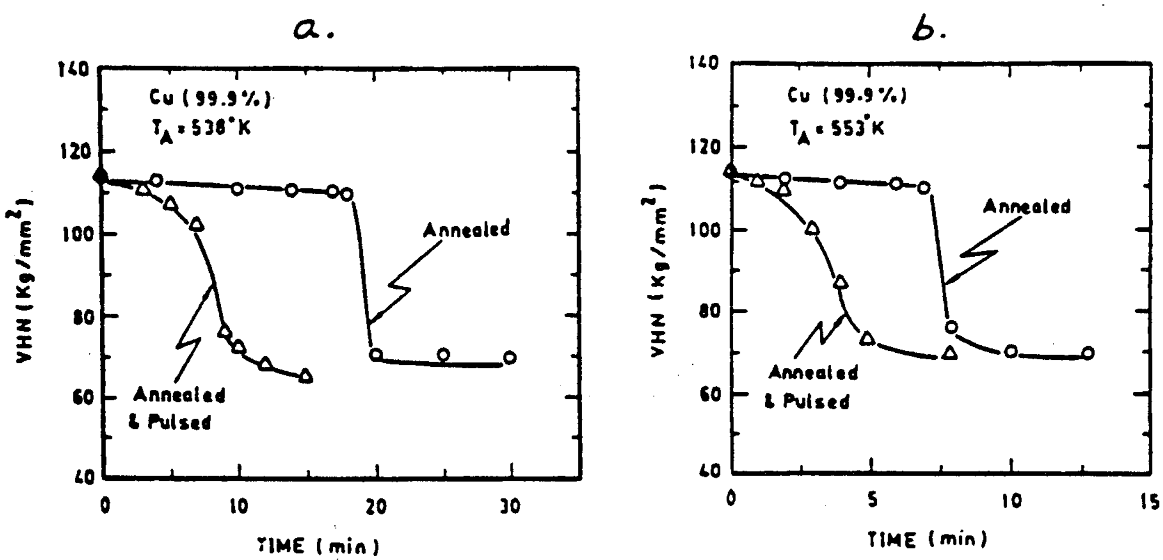


Fig. 6 - Effect of high density ($j = 8 \times 10^4$ A/cm$^2$) electric current pulses ($t_p = 90$ μs and $v_p = 2$ Hz) on the hardness of cold worked (~50% R.A.) copper wire vs annealing time: (a) $T_A = 538$K and (b) $T_A = 553$K.

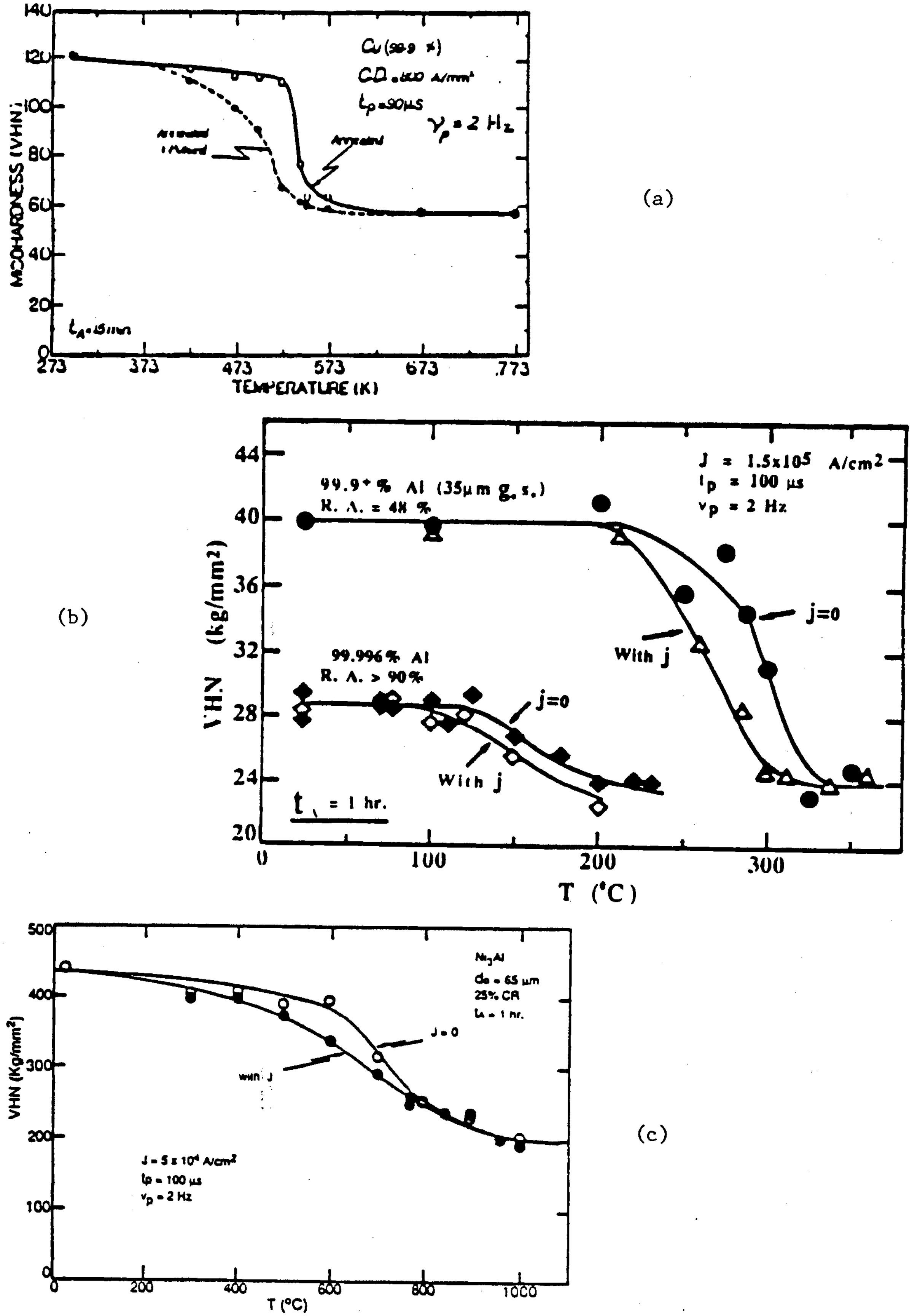


Fig. 7 - Effect of electropulsing on the isochronal annealing of a) Cu, cold work = 50% (8), b) Al annealing time = 1 hr, c) $Ni_3Al$, annealing time = 1 hr, cold work = 25%.

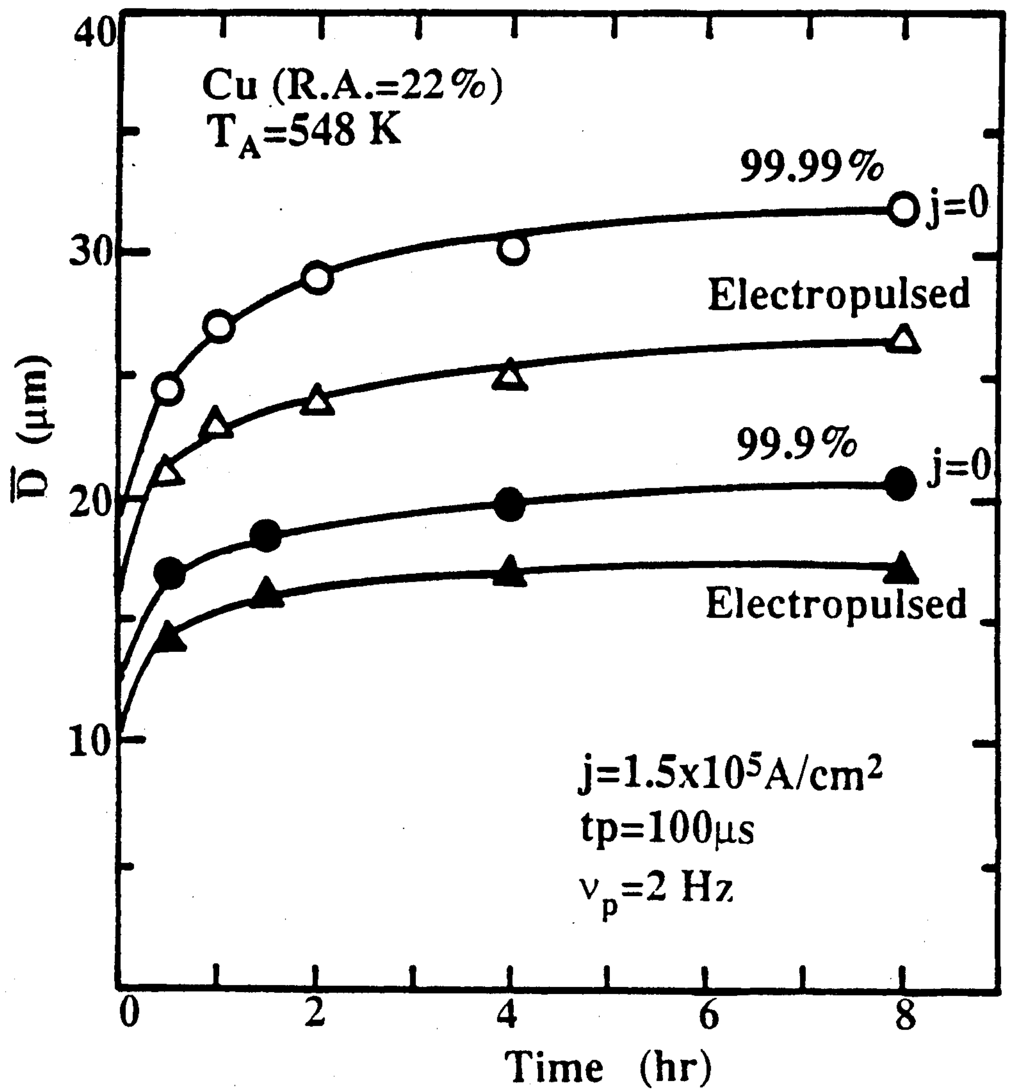


Fig. 8 - Grain growth in Cu as a function of purity and electropulsing.

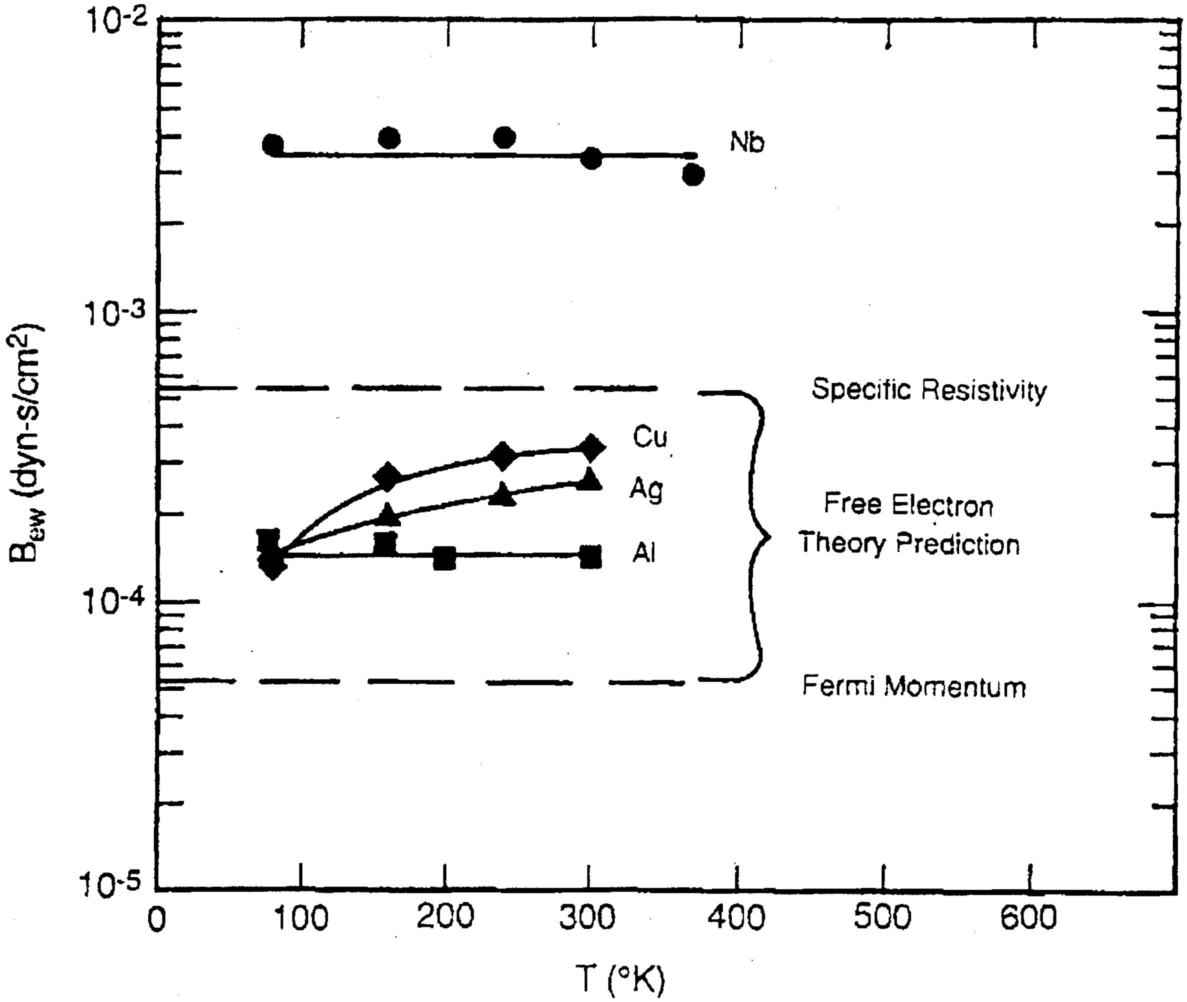


Fig. 9 - Comparison of experimentally-derived values of the electron push coefficient $B_{ew}$ with theoretical predictions based on specific dislocation resistivity and Fermi momentum.

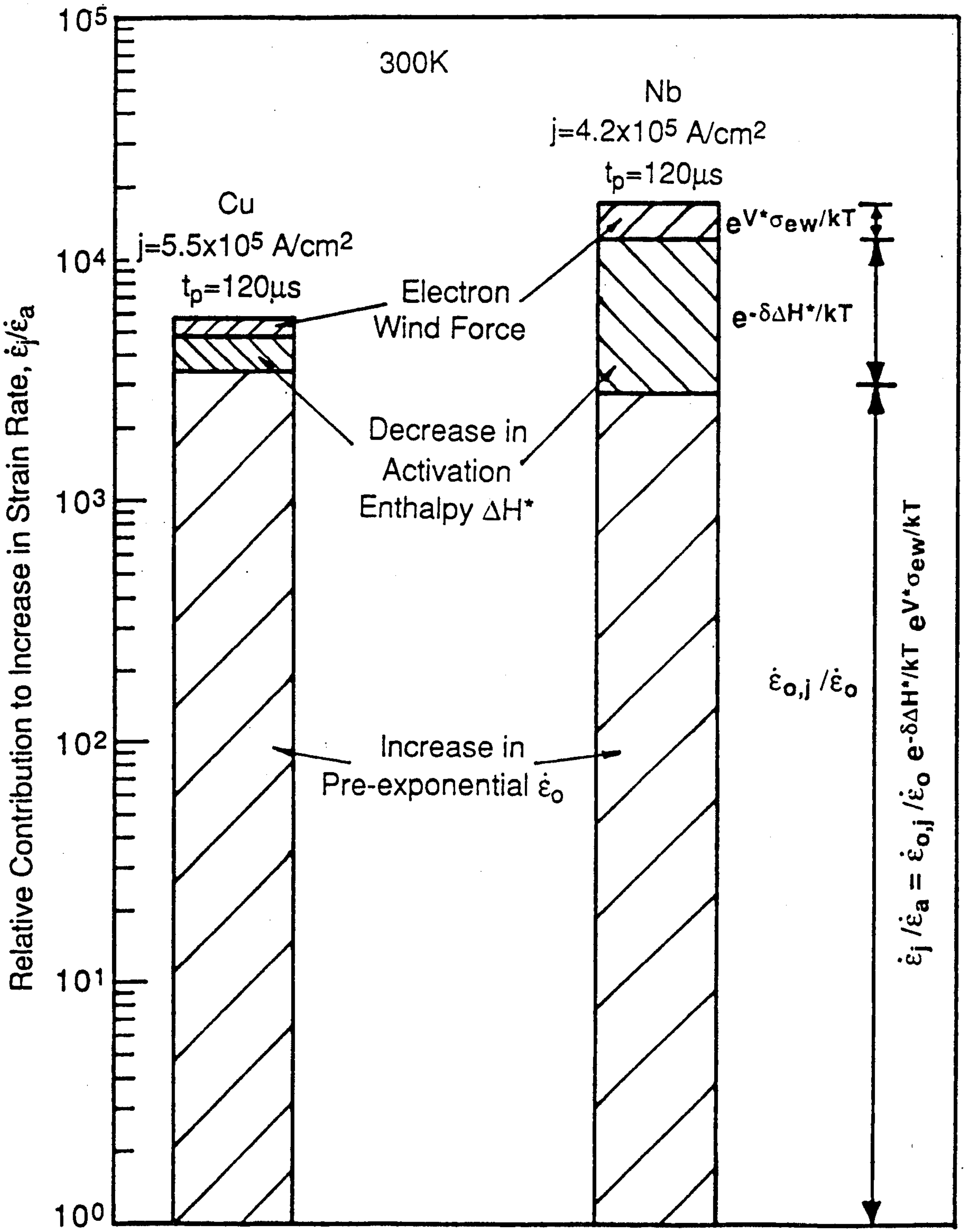


Fig. 10 - Relative contributions of the components of the thermally activated rate equation to the electroplastic effect in FCC Cu and BCC Nb.